Polarization analysis of the magnetic excitations in $Fe_{65}Ni_{35}$ Invar

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Triple-axis inelastic polarized neutron measurements have been performed as a function of temperature on a single crystal of the Invar alloy $Fe_{65}Ni_{35}$ to distinguish longitudinal from transverse magnetic excitations in the magnetically ordered phase. Well below the Curie temperature of 501 K the magnetic excitation spectrum is dominated by conventional transverse spin-wave excitations, which in fact follow the predictions of spin-wave theory very well. In particular, we find no evidence for propagating longitudinal excitations in this system, in sharp contrast to the behavior observed in the amorphous Invar $Fe_{86}B_{14}$ material as well as the non-Invar amorphous system $Fe_{40}Ni_{40}P_{14}B_6$.

For an isotropic ferromagnet the spin-wave dispersion relation in the long-wavelength (small-q) regime is given by[1] $E_{sw}=D(T)q^2$, where D is the spin-wave "stiffness" constant. The general form of the spin-wave dispersion relation, and hence the spin-wave density of states, is the same for all isotropic ferromagnets, while the numerical value of D depends on the details of the magnetic interactions and the nature of the magnetism. For the magnetization, the leading order temperature dependence is given by $M(T)=M(0)(1-BT^{3/2})$, where the coefficient B is simply related in spin-wave theory to D. A measurement of the spin-wave dispersion relation can then be directly related to the bulk magnetization and vice versa. These relationships, as well as many others provided by spin-wave theory, have been found to be in excellent accord with experimental observations for the vast majority of isotropic ferromagnetic materials, with the singular exception of Invar systems.[2–5] In all the Invar materials, whether they be amorphous or crystalline, the relationship between D and B is found to fail in a major way, with the observed stiffness constant D as much as a factor of 2 larger than inferred from magnetization measurements. We previously carried out extensive unpolarized neutron measurements on the amorphous Invar Fe-B system in order to make a detailed comparison between spin-wave theory and experiment.[4] With the exception of the discrepancy between D and B, conventional spin-wave theory was found to work very well in describing the long-wavelength spin dynamics of this system, and thus these unpolarized neutron measurements did not suggest an answer to this problem.

The conventional explanation for this Invar anomaly is that there are additional hidden excitations which participate in reducing the magnetization. If this explanation is correct, then the magnetization and neutron measurements already put stringent conditions on the form that such excitations might take, since there is no freedom to change the *form* of the theory (such as the $T^{3/2}$ behavior for the magnetization, for example). Hence we must have a density of hidden excitations which has precisely the same form as the conventional spin-wave excitations themselves. One possibility which has been suggested[6] is that the (transverse) spin-wave excitations couple to the longitudinal fluctuations, yielding propagating longitudinal excitations which peak at the transverse spin-wave energies. In an unpolarized beam experiment such transverse and longitudinal excitations cannot be distinguished. We therefore carried out inelastic polarized neutron measurements on the $Fe_{86}B_{14}$ Invar system to separate explicitly the longitudinal spin-fluctuation spectrum (S^z) from the usual spin-wave excitations represented by $S^{\pm}=S^x\pm iS^y$, and indeed we observed the presence of longitudinal excitations not only in the vicinity of T_c but substantially below the ordering temperature as well.[7] However, longitudinal excitations were also observed in the non-Invar amorphous ferromagnet $Fe_{40}Ni_{40}P_{14}B_6$, suggesting that these excitations might be related to the amorphous state rather than the Invar anomaly. In our present measurements we in fact do not observe any longitudinal propagating excitations in the crystalline Invar system, and a similar result has been recently obtained in the Fe_3Pt system.[8] Hence the longitudinal excitations observed in $Fe_{86}B_{14}$ are likely not related to the Invar properties but rather have the interesting interpretation that they are unique to the amorphous state. On the other hand, as the Curie temperature is approached from below we do observe longitudinal spin fluctuations, but these fluctuations are diffusive in nature (i.e., they peak at $\Delta E=0$) and appear to be similar to the longitudinal spin diffusion observed in other isotropic ferromagnets near T_c.

The experiments were carried out on the BT-2 triple-axis polarized beam spectrometer at the National Institute of Standards and Technology Research Reactor. Heusler alloy crystals in reflection geometry were employed for both monochromator/analyzer and polarizers. A pyrolytic graphite filter was used to suppress higher-order wavelengths. The sample was a single crystal weighing 22 g, in the shape of a cylinder 1 cm in diameter and 2 cm long. All the present data have been taken in the forward direction, around the (000) reciprocal-lattice point, even though the sample was a single crystal. We used this method so that we might directly compare this system with the data we obtained on amorphous

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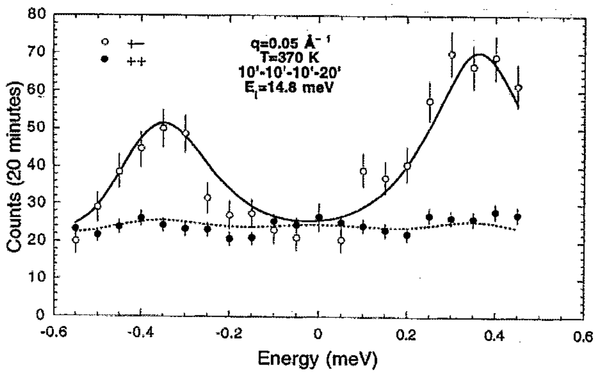

FIG. 1. Spin-flip (○) and non-spin-flip (●) scattering observed in a single crystal of $Fe_{65}Ni_{35}$ (Invar). Clear spin waves are observed in the spin-flip configuration for neutron energy gain ($E<0$) and energy loss ($E>0$), while the very weak inelastic scattering observed in the non-spin-flip configuration is a result of the finite flipping ratio of the instrument.

materials where (000) is the only reciprocal-lattice point, and also in order to be able to apply the field (of 0.6 T) along the cylinder axis to minimize demagnetization effects. In this small-angle regime tight collimation must be employed, and typically we used 20′-10′-10′-20′ (FWHM) in these experiments. We also took some data where the first (in-pile) collimation was 10′ to improve the resolution at smaller wave vectors. The flipping ratio measured through the ferromagnetic sample was between 7 and 13, depending principally on the temperature.

The polarization analysis technique as applied to this problem is in principle straightforward.[9] The (transverse) spin-wave scattering, represented in the Hamiltonian by the raising and lowering operators $S^{\pm}=S^x \pm iS^y$, causes a reversal of the neutron spin. These spin-flip cross sections are denoted by $(+-)$ and $(-+)$. For the results presented here the neutron polarization $\hat{P}$ is perpendicular to the momentum transfer $\mathbf{Q}$, $\hat{P}\perp\mathbf{Q}$, and we may then create a spin wave ($E>0$) or destroy a spin wave ($E<0$) with equal probability. Hence the $(+-)$ and the $(-+)$ cross sections are equal. The longitudinal (S_z) magnetic scattering, on the other hand, is directly related to the non-spin-flip $(++)$ or $(--)$ scattering.

Figure 1 shows the spin-flip and non-spin-flip scattering cross section observed at 375 K for a wave vector of 0.05 Å^{-1}. The scan at this wave vector is restricted in energy to $\sim\pm0.55$ meV due to kinematical constraints of the scattering process.[2] The asymmetry in the spin-wave intensities is due primarily to resolution effects, as discussed in detail in Ref. 10. The solid curve is a fit of the standard spin-wave cross section for isotropic spin waves, convoluted with the instrumental resolution.[10] We obtain excellent agreement with the observations, and for this temperature we have determined that $D=115$ meV Å^2. Also shown in the figure is the non-spin-flip scattering, which is clearly much weaker in strength than the spin-flip scattering. The dashed curve is again a fit, which contains three peaks. One is resolution limited and centered at $E=0$, and at this temperature originates from nuclear incoherent scattering (the scattering intensity of which is q-independent) as well as some magnetic disorder scattering. There are also weak peaks at the spin-wave positions, the intensities of which are approximately $\frac{1}{10}$ of the spin-flip peaks. However, some scattering intensity is expected here due to "leakage" from the imperfect polarization efficiency of the instrument and sample. Indeed the solid curve is the resulting calculated intensity based on the measured spin-flip spin-wave cross section, and clearly provides a complete explanation for this scattering. Thus we conclude that there are no observable longitudinal propagating excitations in this system. The absence of intrinsic peaks in the $(\pm\pm)$ inelastic scattering is in sharp contrast to the behavior observed in the amorphous systems,[7] where we found that the ratio of spin-wave to longitudinal scattering was 2.5:1. Hence it is clear that the longitudinal propagating excitations observed in those systems must have a different origin and are not related to the Invar anomaly.

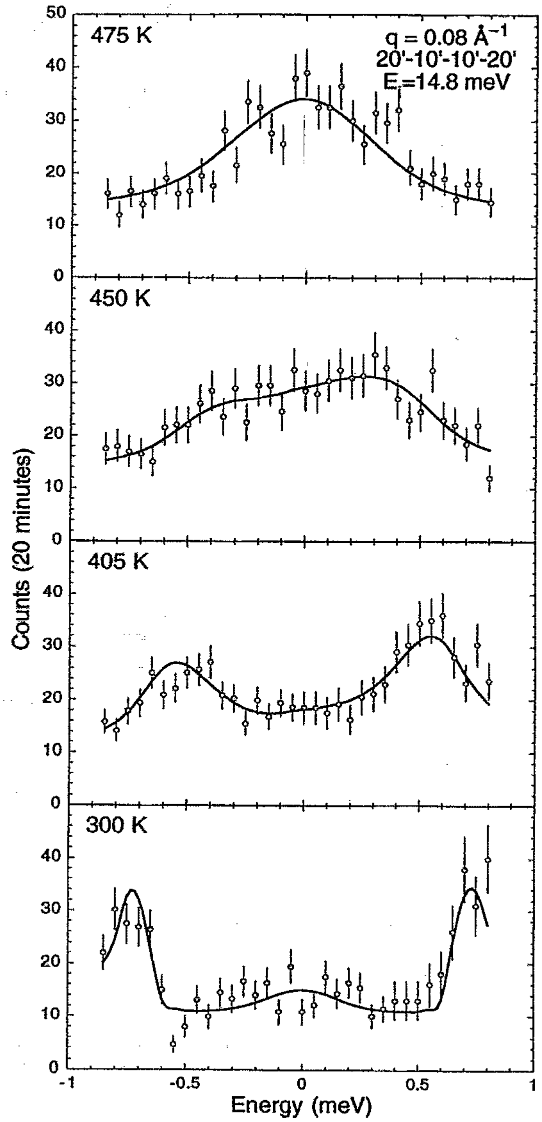

FIG. 2. Temperature evolution of the spin-flip scattering at a wave vector of 0.08 Å^{-1}. The spin waves are seen to renormalize to lower energies and broaden as T_c is approached, as expected.

The temperature evolution of the spin-flip (spin-wave) scattering for $q=0.08$ Å^{-1} is shown in Fig. 2. The solid

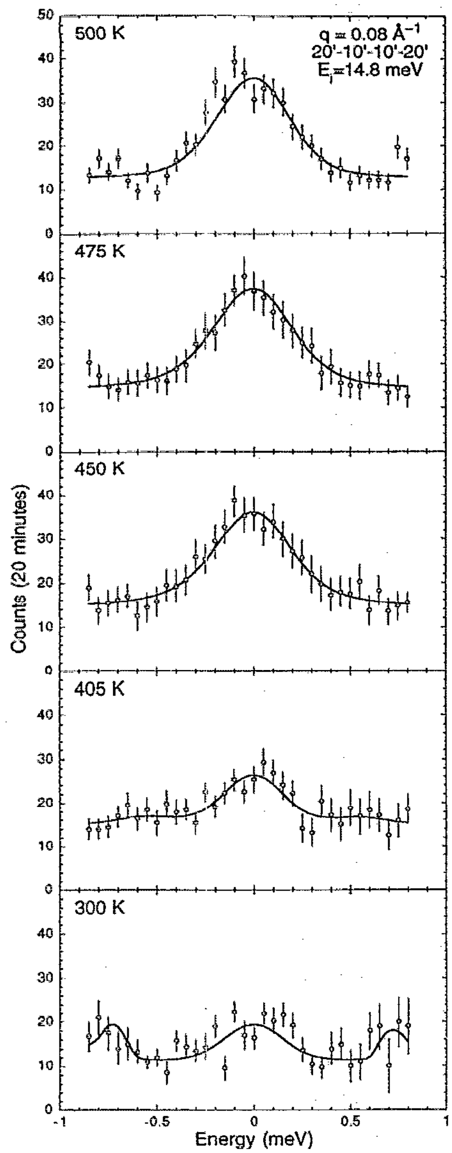

FIG. 3. Temperature evolution of the non-spin-flip scattering at the same wave vector as shown in Fig. 2. The intensity scales are the same so that the data may be directly compared. At room temperature the scattering consists of a resolution-limited elastic peak (nuclear incoherent plus magnetic disorder scattering) plus spin-wave "leakage" due to the imperfect polarization of the instrument and sample. As T_c is approached, a broadened quasielastic magnetic response is observed.

curves again represent the least-squares fits to the data of the spin-wave cross section convoluted with the instrumental resolution and polarization.[10] At room temperature we see two well-separated spin-wave peaks, and as T_c (501 K) is approached the spin waves renormalize to lower energy and broaden as expected. At 475 K we find that the spin waves are almost overdamped, and in fact after convolution with the instrumental resolution only a single broad peak is observed at this q. These data can be compared directly with the longitudinal (non-spin-flip) scattering as shown in Fig. 3. At room temperature the scattering consists of a resolution-limited elastic peak plus spin-wave peaks from "leakage." As the temperature is increased towards T_c the spin-wave peaks are diminished because of the improvement in the flipping ratio at higher temperatures and also because the spin waves are broadened. What is clear from these data is that a quasielastic peak develops as T_c is approached. With the assumption that the intrinsic cross section is a Lorentzian we obtain an intrinsic (half) width $\Gamma \approx 0.07$ meV at this q.

This quasielastic scattering originates from simple spin diffusion.[6] For $T \geqslant T_c$, x, y, and z are equivalent for an isotropic ferromagnet by definition, while below T_c this symmetry is broken and we have a transverse (x,y) and longitudinal (z) susceptibility. In the hydrodynamic regime above T_c only spin diffusion occurs, and it is reasonable to expect that the longitudinal response will remain diffusive below T_c.[11] Hence we do not believe that this scattering is related to the Invar effect but rather is the expected behavior for an isotropic ferromagnet for $T \lesssim T_c$. This still leaves the origin of the Invar anomaly as a mystery, and also implies that the longitudinal excitations observed in the amorphous ferromagnets are a new phenomenon. Both of these effects warrant further investigation.

The research at Maryland was supported by the NSF, Grant No. DMR 93-02380.

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